

In the Specification

Please replace the paragraph spanning pages 2 and 3 with the following:

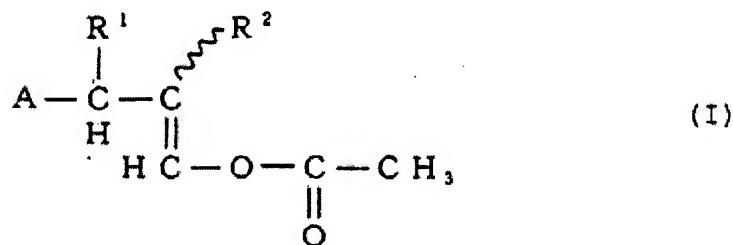
SUMMARY OF THE INVENTION

~~An object of the present invention is to~~ We provide a process for producing a 1-acetoxy-3-(substituted phenyl)propene compound useful, as an intermediate material, for perfumes, pharmaceuticals, agricultural chemicals and other organic synthetic chemicals with high efficiency and an easy process.

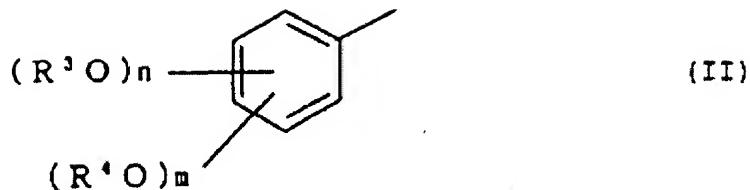
On page 3, please delete the first full paragraph.

Please replace the paragraph spanning pages 3 – 5 with the following:

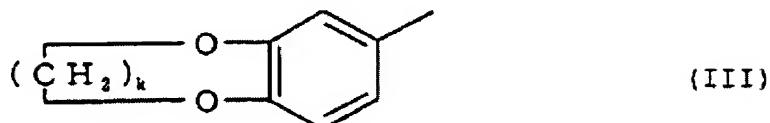
The process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound represented by the general formula (I):



in which formula (I), R¹ and R², respectively and independently from each other, represent a member selected from the groups consisting of a hydrogen atom and alkyl groups having 1 to 10 carbon atoms, R¹ and R² may form, together with carbon atoms located in the 2- and 3-positions of the propene group, a cyclic group; and A represents a member selected from a group of substituted phenyl groups represented by the formulae (II) and (III):

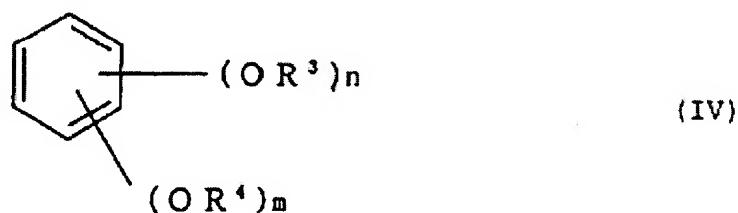


and

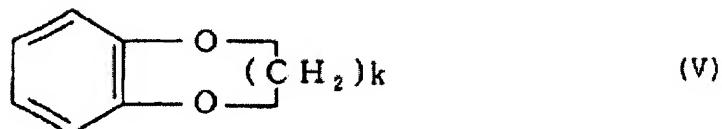


wherein R^3 and R^4 , respectively and independently from each other, represent an alkyl group having 1 to 4 carbon atoms, m represents an integer of 0 or 1 to 4, n represents an integer of 1 or 5 and k represents an integer of 1 or 2,

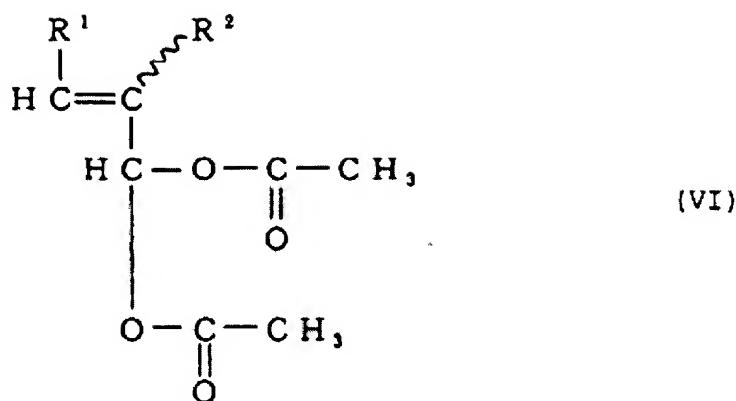
comprises reacting a benzene compound selected from those represented by the general formulae (IV) and (V):



and



in which formula (IV) and (V), R^3 and R^4 and n , m and k are as defined above, with an alkenylidene diacetate compound represented by the general formula (VI):



in which formula (VI), R^1 and R^2 are as defined above,

in the presence of a catalyst comprising at least one compound selected from the group consisting of (a) halogenated boron compounds, (b) triflate compounds of Group 11 elements of the Periodic Table, (c) halogenated compounds of Group 12 elements of the Periodic Table, and (d) triflate compounds and halogenated compounds of tin and lanthanoid elements of atomic numbers 58 and 66 to 71.

On page 5, please replace the first five full paragraphs with the following:

~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, the~~ The benzene compounds represented by the formula (IV) ~~is~~are preferably selected from the group consisting of anisole, veratrol, hydroquinone dimethylether, Pyrogallol trimethylether and hydroxyhydroquinone trimethylether.

~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, the~~ The benzene compounds represented by the formula (V) ~~is~~are preferably selected from the group consisting of 1,2-methylenedioxybenzene and 1,2-ethylenedioxybenzene.

~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, the~~ The alkenylidene diacetate is preferably selected from the group consisting of

3,3-diacetoxy-2-methylepropene, 3,3-diacetoxy propene, 3,3-diacetoxy-1-methylpropene, 3,3-diacetoxy-2-ethyl propene, 3,3-diacetoxy-1-ethylpropene, and 3,3-diacetoxy-1-ethyl-2-methyl-propene.

~~In the process of the present invention for producing a 1-acetoxy 3-(substituted phenyl)propene compound, the~~ The reaction is preferably carried out in a molar ratio of the benzene compound to the alkenylidene diacetate compound of 1:1 to 50:1.

~~In the process of the present invention for producing a 1-acetoxy 3-(substituted phenyl)propene compound, the~~ The catalyst is preferably present in an amount of 0.005 to 1 mole per mole of the alkenylidene diacetate compound.

Please replace the paragraph spanning pages 5 and 6 with the following:

~~In the process of the present invention for producing a 1-acetoxy 3-(substituted phenyl)propene compound, the~~ The halogenated boron compounds (a) usable for the catalyst are preferably selected from boron fluorides, boron trifluoride-diethylether complexes, borontrifluoride-tetrahydrofuran complexes, boron trifluoride-acetic acid complex salt, boron trifluoride dehydrate, and boron trifluoride-n-butylether complexes.

On page 6, please replace the first four full paragraphs with the following:

~~In the process of the present invention for producing a 1-acetoxy 3-(substituted phenyl)propene compound, the~~ The triflate compounds (b) of Group II elements of the Periodic Table usable for the catalyst are preferably selected from the group consisting of copper triflate and silver triflate.

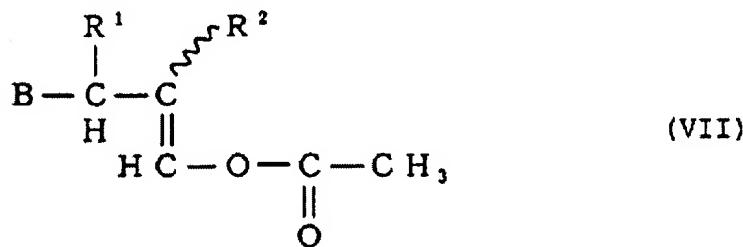
~~In the process of the present invention for producing a 1-acetoxy 3-(substituted phenyl)propene compound, the~~ The halogenated compounds (c) of Group 12 elements of the Periodic Table usable for the catalyst are preferably selected from the group consisting of zinc fluoride, zinc chloride, zinc bromide, zinc iodide, cadmium fluoride, cadmium chloride, cadmium bromide, cadmium iodide, hydrogen fluoride, mercury chloride, mercury bromide, and mercury iodide.

~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propane compound, the~~ The triflate and halogenated compounds (d) of tin and lanthanoid elements of atomic numbers 58 and 66 to 71 are preferably selected from the group consisting of triflates, fluorides, chloride, bromides, and iodide of tin, cerium, dysprosium, holmium, erbium, thulium, ytterbium and lutetium.

~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propane compound, the~~ The reaction is preferably carried out in an atmosphere consisting of a non-reactive gas to the above-mentioned compounds of the formulae (IV), (V) and (VI), the above-mentioned catalyst and the resultant reaction products.

Please replace the paragraph spanning pages 6 and 7 with the following:

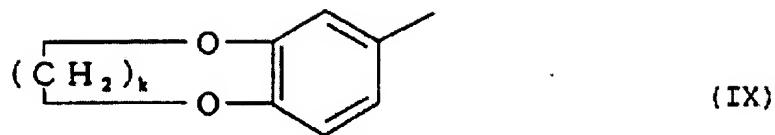
~~In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propane compound, the~~ The compounds of the formula (I) are preferably selected from the compounds represented by the general formula (VII):



In which formula (VII), R¹, R² are as defined above, B represents a member selected from a group of substituted phenyl groups represented by the formulae (VIII) and (IX):



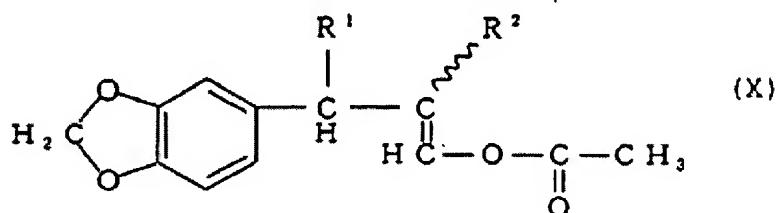
and



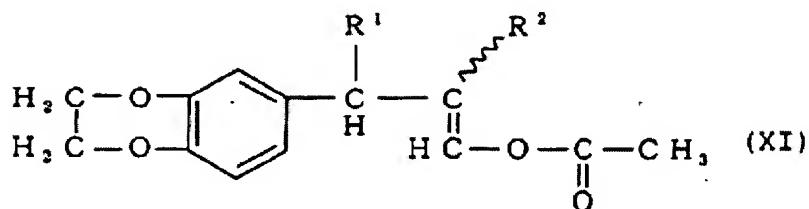
In which formulae (XVIII) and (IX), R³ and R⁴ and k are as defined above.

Please replace the paragraph spanning pages 7 and 8 with the following:

In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, the The compound of the formula (I) is preferably selected from 1-acetoxy-3-(3,4-C1 to C2 alkylene dioxypyhenyl)propenes represented by the formulae (X) and (XI):



and



On page 8, please replace the second and third full paragraphs with the following:

In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, preferably Preferably in the formulae (X) and (XI), R¹ represents a hydrogen atom and R² represents a methyl group.

In the process of the present invention for producing a 1-acetoxy-3-(substituted phenyl)propene compound, the The compound of the formula (I) is preferably selected from the groups con-

sisting of 1-acetoxy-2-methyl-3-(3,4-methylenedioxyphenyl)propene, 1-acetoxy-2-methyl-3-(3,4-ethylenedioxyphenyl)propene, 1-acetoxy-2-methyl-3-(4-methoxyphenyl)propene, 1-acetoxy-2-methyl-3-(2,5-dimethoxyphenyl)propene, 1-acetoxy-2-methyl-3-(3,4-dimethoxyphenyl)propene.

On page 8, please replace the fifth paragraph with the following:

The Periodic Table used in the present invention is based on the 18 Groups-Type Elemental Periodic Table, IUPAC and Nomenclature in Inorganic Chemistry, 1990 Rule.

On page 9, please replace the first three paragraphs with the following:

~~BEST MODE FOR CARRYING OUT THE INVENTION~~ DETAILED DESCRIPTION

The 1-acetoxy-3-(substituted phenyl)propene compound produced by the process of the present invention is represented by the above-mentioned general formula (I) and includes a plurality of types of stereoisomers due to asymmetric carbon atoms and/or a double bond contained in the molecule of the compound.

The process of the present invention for producing 1-acetoxy-3-(substituted phenyl)propene compound comprises the step of reacting at least one member selected from a group of benzene compounds represented by the above-mentioned general formulae (IV) and (V) with an alkenylidene diacetate represented by the general formula (VI) in the presence of a specific catalyst which will be illustrated in detail hereinafter. The benzene compounds represented by the formulae (IV) and (V) correspond to the substituted phenyl groups represented by the general formula (II) and (III), and the alkenylidene diacetate of the general formula (VI) corresponds to a 1-acetoxypropene group bonded to the A group contained in the general formula (I).

The specific catalysts for the process of the present invention comprises at least one member selected from the group consisting of:

(a) halogenated boron compounds,

- (b) triflate compounds of Group II elements of the Periodic Table,
- (c) halogenated compounds of Group 12 elements of the Periodic Table, and (d) triflate compounds and halogenated compounds of tin and lanthanoid elements of atomic numbers 58 and 66 to 71.

Please replace the paragraph spanning pages 9 and 10 with the following:

In the process ~~of the present invention~~, the benzene compound represented by the general formula (IV) is preferably selected from anisole, veratrole, hydroquinonedimethylether, pyrogalloltrimethylether, and hydroxyl hydroquinonetrimethylether. Among them, anisol and veratrol are particularly preferably used. These compounds may be of a common trade grade.

Please replace the paragraph spanning pages 10 and 11 with the following:

The halogenated boron compound (a) for the catalyst usable for the process ~~of the present invention~~ includes, for example, boron fluoride, boron trifluoride-diethylether complex, boron trifluoride-tetrahydrofuran complex, boron trifluoride-acetic acid complex salt, boron trifluoride-dihydrate and boron trifluoride-n-butylether complex. Among them, boron trifluoride-ether complex and boron trifluoride-acetic acid complex salt are more preferably employed. These compounds may be of a trade grade.

Please replace the paragraph spanning pages 11 and 12 with the following:

In the process ~~of the present invention~~, the catalyst is preferably employed in an amount of 0.005 to 1 mole, more preferably 0.01 to 0.5 mole, still more preferably 0.01 to 0.2 mole, per mole of the alkenylidene diacetate. If the catalyst is used in an amount of more than 1 mole, complicated procedures may be needed for recovery, decomposition and disposal of the catalyst after the reaction is completed, and may cause the practice of the process ~~of the present invention~~ in the industrial

scale to be inconvenient. Also, if the amount of the catalyst is less than 0.005 mole, the reaction may not be completed within a practical time, for example, within 24 hours.

On page 12, please replace the first two full paragraphs with the following:

The reaction in the process ~~of the present invention~~ may be carried out in a solvent medium. Usually, the reaction is preferably not carried out in a solvent medium. For the solvent, aromatic hydrocarbons, for example, benzene and toluene, xylene; halogenated aromatic hydrocarbons, for example, chlorobenzene; and halogenated aliphatic hydrocarbons, for example, methylene chloride and dichloroethane, may be employed.

The reaction temperature for the process ~~of the present invention~~ can be appropriately established in response to the types and concentrations of the starting compounds and catalysts. Usually, the reaction is carried out at a temperature of -10 to 80°C, more preferably 0 to 60°C. The reaction time for the process ~~of the present invention~~ can be appropriately established in consideration of the types and concentrations of the starting compounds and catalysts and the reaction temperature. Usually, the reaction time is preferably in the range of from 0.5 to 24 hours, more preferably 0.5 to 12 hours.

Please replace the paragraph spanning pages 12 and 13 with the following:

There is no specific limitation to the type of the reaction atmosphere for the process ~~of the present invention~~. Usually, the reaction of the process ~~of the present invention~~ is carried out in a gas nonreactive to the starting compounds (namely, the compounds of the general formulae (I) and (II), the catalyst and the resultant products, for example, a gas atmosphere or flow comprising at least one gas selected from nitrogen gas and inert gases, for example, argon gas. The reaction is usually carried out at the ambient atmospheric pressure. However, the reaction pressure is not limited to that mentioned above.

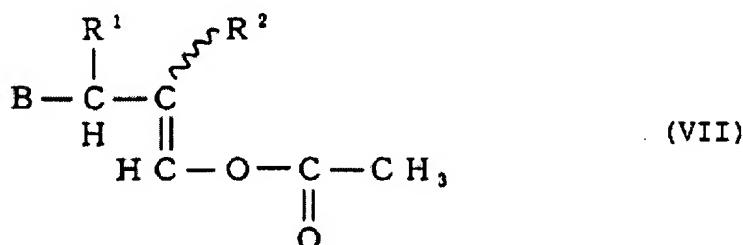
On page 13, please replace the first two full paragraphs with the following:

The 1-acetoxy-3-(substituted phenyl)propene compound produced in accordance with the process ~~of the present invention~~ is usually refined by separating the compound from the resultant reaction mixture liquid after the reaction is completed by a usual separate-recovery procedure, for example, an extraction, a concentration and a filtration and then optionally by applying a refining procedure, for example, a distillation, recrystallization and various chromatographies, to the separate-recovered fraction.

In the general formula (I) representing the 1-acetoxy-3-(substituted phenyl)propene compound produced by the process ~~of the present invention~~, R¹ and R² represent a hydrogen atom or a C1 – C10 alkyl group, and preferably, at least one of R¹ and R² represents a C1 – C10 alkyl group. The C1 – C10 alkyl groups represented by R¹ and R² include methyl, ethyl, propyl, butyl, pentyl, hexyl, heptyl, octyl, nonyl and decyl groups. These groups respectively include a plurality of isomers. The alkyl group represented by R¹ and R² is preferably a methyl group.

Please replace the paragraph spanning pages 14 and 15 with the following:

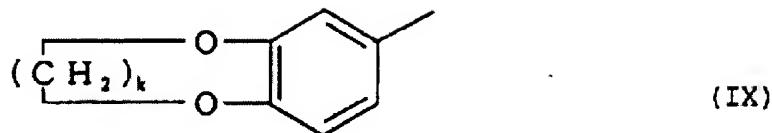
Among the compounds of the general formula (I) produced by the process ~~of the present invention~~, the 1-acetoxy-3-(substituted phenyl)propene compounds represented by the general formula (VII):



In which formula (VII), R¹, R² are as defined above, B represents a member selected from a group of substituted phenyl groups represented by the formulae (VIII) and (IX):



and



In which formulae (XVIII) and (IX), R³ and R⁴ and k are as defined above, are novel compounds.

On page 15, please replace the first full paragraph with the following:

In the case where the A in the general formula (I) represents the substituted phenyl groups of the general formula (III), the 1-acetoxy-3-(substituted phenyl)propene compounds of the present invention represented by the general formula (I) is preferably selected from the 1-acetoxy-3-(3,4-C1-C2alkylenedioxy-phenyl)propenes represented by the general formulae (X) and (XI). In this case, in the general formulae (X) and (XI), preferably, R¹ represents a hydrogen atom and R² represents a methyl group.

On page 15, please replace the third and fourth paragraphs with the following:

Accordingly, the 1-acetoxy-3-(substituted phenyl)propene compounds of the present invention represented by the general formula (I) are preferably selected from the group consisting of 1-acetoxy-2-methyl-3-(3,4-methylenedioxyphenyl)propene, 1-acetoxy-2-methyl-3-(3,4-ethylene-diox-phenyl)propene, 1-acetoxy-2-methyl-3-(4-methoxyphenyl)propene, 1-acetoxy-2-methyl-3-(2,5-dimethoxyphenyl)propene, and 1-acetoxy-2-methyl-3-(3,4-dimethoxyphenyl)propene.

EXAMPLES

~~The present~~Selected aspects of the invention will be further illustrated by the following examples which are not intended to limit the scope of the ~~present invention~~ appended claims in any way.

On page 31, please replace the third full paragraph with the following:

~~INDUSTRIAL APPLICABILITY OF THE INVENTION~~

The process ~~of the present invention~~ enables a 1-acetoxy-3-(substituted phenyl)propene compound useful, as an intermediate, for perfumes, pharmaceutical chemicals, agricultural chemicals and other organic synthetic chemicals, to be easily produced in a high yield. Thus the process ~~of the present invention~~ for producing 1-acetoxy-3-(substituted phenyl)propene compounds has a high applicability in industry. Also, the 1-acetoxy-3-(substituted phenyl)propene compounds produced by the process ~~of the present invention~~ include new compounds.